A New Approach to Grow GaN by Low-Pressure MOCVD Using a Three Steps Technique

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A new growth approach has been investigated to grow GaN on sapphire substrate using low-pressure metalorganic chemical vapor deposition (LP-MOCVD). This technique consists of first depositing an AlN thin film by atomic layer epitaxy (ALE) and subsequent application of the conventional two steps method. This three steps technique reveals to be a promising issue to produce good quality GaN epilayers.

1. Introduction

III-nitride semiconducors demonstrated a great potential for the fabrication of optoelectronic devices operating in the ultraviolet and visible spectral region. This is due in part to the growth of high quality GaN epilayers using metalorganic chemical vapor deposition (MOCVD). Besides the conventional and widely used two-steps technique, several other groups investigated new approaches to deposit the nucleation layer, as it is one of the key factors determining the subsequent epilayer properties.

Recently our group reported the possibility of controlling the polarity of GaN by inserting a few monolayers of aluminum between the nitrided sapphire substrate and the conventional low-temperature buffer layer [1)]. In order to further improve the crystallinity of the epilayers we have developed a new three steps procedure that requires first the deposition of AlN using atomic layer epitaxy (ALE). In the present work we report the first ALE of AlN directly on sapphire using LP-MOCVD and demonstrate the capability of the three steps technique to grow GaN exhibiting a smooth morphology and good structural properties as well. The results suggest that AlN grown by ALE is an efficient alternative way to improve the quality of heteroepitaxy and ensure a better \`extraction\` of the crystallographic information form the substrate.

2. Experimental

GaN epilayers were grown on (0001) sapphire substrates at 1080℃ in a horizontal three-gas flow injection LP-MOCVD reactor. The precursor materials used were trimethylgallium (TMG), trimethyl-aluminum (TMA) and ammonia (NH_3) while H_2 was used as a carrier gas. The detailed growth procedure is described in Fig.1. After thermal cleaning in H_2 ambient at 1100°C, the substrates were nitrided for 90 sec at the same temperature and under a pressure of 76 torr. The ALE mode growth of the AlN film then proceeded by supplying alternatively TMA and NH_3. One whole growth cycle required 8 sec time while the sweep-out interval between the two source materials pulses was fixed to 2 sec. The substrate temperature is then decreased to 550 ^{o}C to expose the sample surface to a TMAl pre-flow for 7 sec, followed by the deposition of a 20-nm-thick GaN buffer layer. Finally the temperature is raised to 1080 ^{o}C to grow 3 μm-thick GaN films. During these last three steps the reactor pressure was set to 200 Torr.

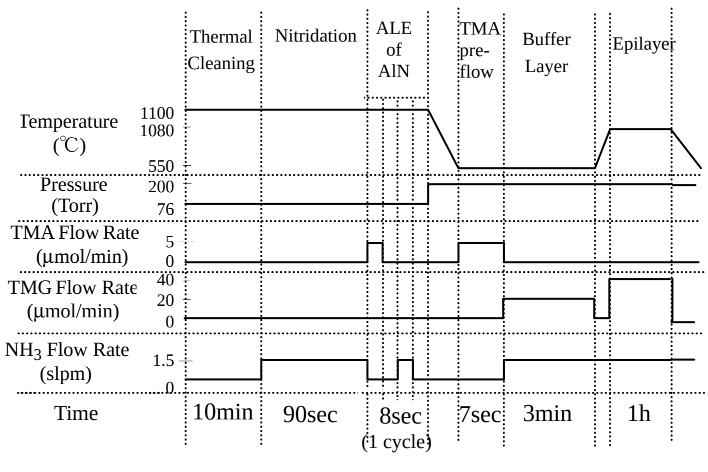

Fig. 1. GaN growth sequences and flow patterns of the precursor materials. The number of cycles in the ALE step is set depending on the AlN film thickness to be grown.

The samples were analyzed using a high-resolution 4-crystals X-ray diffractometer and a Normarski optical microscope. Moreover, a spectroscopic phase-modulated ellipsometry system mounted on the MOCVD reactor also allowed the implementation of an in-situ real time diagnosis of the growth process.

3. Results and Discussion

Figure 2 shows the X-ray rocking curve (XRD) recorded for the (002) reflection of a relatively thick AlN grown directly on nitrided sapphire by ALE. The number of cycle was set to 400. One cycle consisted successively of 2sec TMA supply, 2sec purge, 2sec NH_3 supply and 2sec purge. In the absence of self-limitation process a TMA flow rate of 5 µmol/min was chosen in order to promote the deposition of one AlN monolayer/cycle. The ellipsometric signals showed clearly the step growth while the (002) XRC FWHM value of 140 arcsec is indicative of a well-oriented initial columnar film structure.

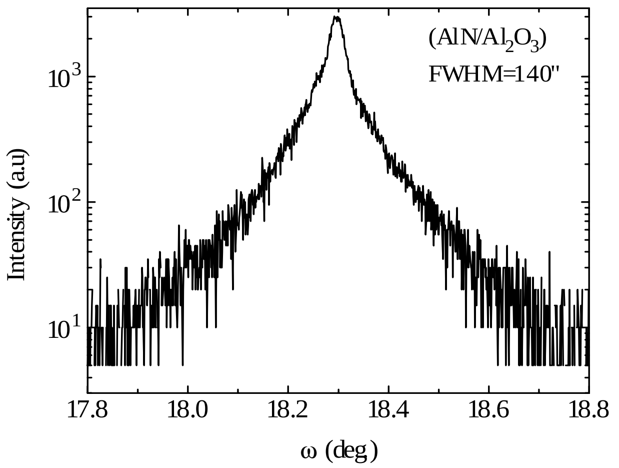

Fig. 2. (002) X-ray rocking curve of AlN film grown directly on nitrided sapphire substrate using ALE and 400 cycles.

To investigate the capability of the proposed three steps technique, the effects of two important parameters were actually studied, namely AlN thickness and TMA pre-flow time. In this purpose GaN samples were grown using 7 sec (#0181, #0179 and #188) and 5 sec (#0175) TMA pre-flow time. Figure 3 displays the variation of the symmetric and asymmetric XRC FWHMs as a function of the number of cycles used for AlN-ALE. One could notice that sample #0179, which corresponds to 5 cycles, was found to exhibit the best properties with respect to both the surface smoothness (Ga-polar) and crystallinity. These results are very significant if one consider the data recorded for both the

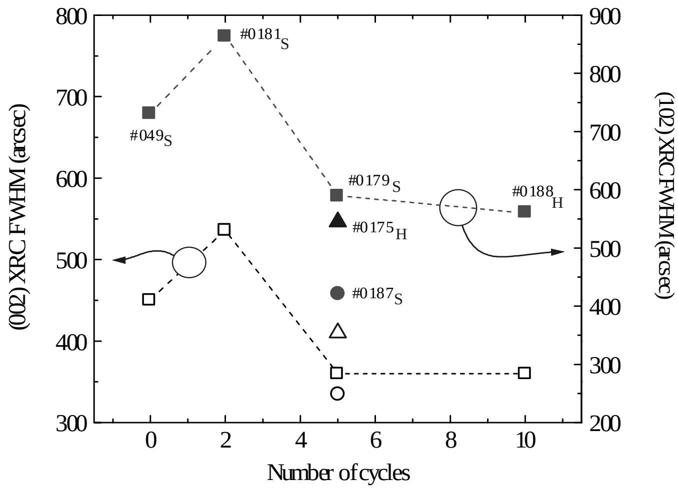

Fig.3. Effect of the AlN film thickness on the structural properties of GaN epilayers. Samples #0049 and #0175 were grown using a TMA pre-flow time of 5sec while sample #0187 is ~7µm-thick. The characters S and H refer to a smooth and hexagonal surface morphology, respectively.

reference sample #049 grown using (0 cycle and 5 sec TMA pre-flow time) and sample #0175. Indeed the former shows higher values of XRC FWHMs, while the latter was characterized by a hexagonal pyramidal morphology, i.e. N-polar. The improvement in the structural properties is believed to result from a reduction of tilting and twisting in the bottom layer. More recently very interesting XRC FWHM data were measured for a sample grown during 3 hours to a thickness of ~7 µm (sample #187), that is to say 335 and 421 arcsec for the GaN (002) and (102) diffraction planes, respectively. However to assess the origin of such improvement (epilayer low growth rate or epilayer thickness) further work is needed. To complete the present study optical and electrical characterization of the samples are also under way.

4. Conclusion

A new three steps growth technique that combines successively the deposition of high quality AlN using ALE, subsequent exposition of the sample to a TMA pre-flow and the application of the conventional 2 steps growth method, has been used successfully to grow device quality GaN in a LP-MOCVD reactor. The preliminary results showed that the morphology and structural properties of the GaN epilayers were strongly dependent on the AlN thickness as well as the TMA pre-flow time.

1) D. H. Lim, Y. Taniyasu, S. Arima, K. Susuki, B. L. Liu, H. J. Lee and Y. Yoshikawa, Proc. of the 2000 Elec. Mater. Conf., June 21-23, 2000, Denver, Colorado (USA).